Pair correlated atoms with a twist

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We present an analysis of the quantum state resulting from the dissociation of diatomic molecules prepared in a condensate vortex state. The many-body state preserves the rotational symmetry of the system in quantum correlated states by having two equally populated components with angular momentum adding to unity. A simple two-mode analysis and a full quantum field analysis is presented for the case of non-interacting atoms and weak depletion of the molecular condensate.

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I. INTRODUCTION

Since the 1995 experiments with the first production of atomic Bose-Einstein condensates, degenerate quantum gasses have constituted a very active field of research. A wide variety of means exists for detection and control of the properties of these systems, and the early works have been followed by progress on degenerate fermionic systems, on mixtures of different species, and on conversion between atomic and molecular quantum gasses.

The coherence properties of the degenerate quantum states have been verified implicitly by measurements of the response properties of the systems and explicitly by the observation of robust interferences [1] and topological structures [2]. Recently, a vortex lattice in a system of fermionic atoms was shown to extend over the BCS-BEC crossover towards formation of bosonic diatomic molecules on the molecular side of a field controlled Feschbach resonance [3].

Many properties of condensates are well described by mean field theories and the Gross-Pitaevskii equation, but in some cases mean field theories may completely fail and totally forbid processes, which occur peacefully according to a full quantum state analysis. Important examples of such processes are the period-doubling observed in a shaken lattice [4] and the break-up of a moving condensate in a lattice due to four wave mixing [5]. Both processes are driven by collisions of pairs of condensate atoms which emerge in two new momentum states. Within a mean field theory, these processes, like the equivalent parametric amplification process in optical down-conversion and four wave mixing will only get initiated if a non-vanishing mean field with the final state character is seeded to the solution of the Gross-Pitaevskii equation [6]. From a formal perspective, the fact that only the sum of the phases of the two final state components and not the individual phases are locked to the initial state wave functions causes the fields to remain in the vacuum state in order not to break the phase symmetry. If the physical problem is simple, e.g., in the few-mode quantum optical problems, one may alternatively have recourse to a full quantum many-body theoretical analysis. In the context of quantum gasses, we have previously [7] studied the "degenerate down-conversion" of a molecular condensate by dissociation to a single trapped atomic condensate in such a full quantum treatment and shown that despite the absence of an atomic condensate phase, the atomic component does indeed accumulate in a single preferred quantum state. Dissociation into two atomic beams have smilarly been proposed as a means for generation of Einstein-Podolsky-Rosen correlations [8].

Note that in four-wave mixing processes a mean-field solution not only breaks the phase symmetry of the field, it also breaks the translational invariance of the problem as dramatically observed as period doubling in [4], and likewise the different populated momentum components in [5] will have spatial interference patterns with periods exceeding the one of the lattice potential used in the experiments. In this paper, we consider a process where the symmetry breaking is similarly spectacular, namely the down conversion by photo dissociation of a molecular condensates which is prepared in a single vortex state. In a cylindrically shaped trap, a vortex state is a topologically stable state of a quantum gas with a vanishing density along the cylinder axis and a phase which changes by 2π as one follows a closed loop around the axis. The rotational symmetry around the z axis is a fundamental property of the many-body Hamiltonian and both mean field solutions and more elaborate theories must respect this symmetry. Microscopically, the vortex solution is consistent with a Hartree product state description with a product of single particle $M = 1$ eigenstates of the azimuthal angular momentum. In this state, every molecule has a unit angular momentum around the condensate axis, which cannot be transferred to a pair of atoms populating the same ($m = 1/2$?) quantum state. Conversely, any mean field solution for the atomic system will necessarily correspond to an integer angular momentum per atom, and would hence imply an even angular momentum of every pair, i.e, of the molecule.

The molecules must dissociate to at least two different atomic states with azimuthal quantum numbers which add to the molecular value, $m + m' = M$. The easiest situation to deal with is one where only two states with

angular momentum $m = 0$ and $m' = 1$ get populated. We shall address this case in Sec. II. In particular we shall discuss to which extent a strongly number correlated quantum state of this kind state is distinguishable from a state which macroscopically populates an even weight superposition of the same two single particle states. The restriction to only two finally occupied states should be justified by a more elaborate treatment of the many-body Hamiltonian, and in Sec. III, we model the actual process in which the atoms are not put directly into two states extending over vast and partly non-overlapping regions of the atomic single particle quantum states, but where the molecular dissociation enforces a localized common origin of the matter waves. The problem is solved exactly by a Bogoliubov tranformation, and we identify different regimes and the validity of the few-mode Ansatz.

II. TWO-MODE ANALYSIS

For simplicity we ignore interaction between atoms, between molecules, and between atoms and molecules. We also make the assumption that only one quantum state for the molecules and two quantum states for the atoms are relevant. The molecule state should be the macroscopically occupied state of the molecular condensate. The two atomic states are assumed to be selected by a resonance condition: They could for example be the two lowest 2D single particle eigenfunctions in a harmonic trap [cf. Eq. (11) below], $\Phi_{00}(x,y) \propto \exp(-\rho^2/2a_{\rm osc}^2), \Phi_{01}(x,y) \propto (x+iy)\exp(-\rho^2/2a_{\rm osc}^2)$, where $\rho^2 = x^2+y^2$, and where the oscillator length $a_{\rm osc} = \sqrt{\hbar/m_{\rm atm}\omega}$ is defined in terms of the atomic mass $m_{\rm atm}$ and the trap frequency ω. The lowest state Φ_{00} has angular momentum $m = 0$, while Φ_{01} has $m' = 1$ so that they fulfill the condition for angular momentum conservation when molecules with $M = 1$ are dissociated, $m + m' = M$.

If we can drive the system exactly at resonance, i.e., either the atomic and molecular states are degenerate, or in a laser induced dissociation process, the field frequencies involved exactly fulfill the Bohr frequency condition for quantum transitions, our model Hamiltonian becomes

$$\hat{H} = \beta\, \hat{c}\hat{a}_0^\dagger \hat{a}_1^\dagger + \beta^*\, \hat{c}^\dagger \hat{a}_0 \hat{a}_1. \tag{1}$$

Here β quantifies the strength of the microscopic coupling, the operator $\hat{c}(\hat{c}^\dagger)$ annihilates (creates) a molecule, and the operators $\hat{a}_0, \hat{a}_1(\hat{a}_0^\dagger, \hat{a}_1^\dagger)$ annihilate (create) atoms in the two atomic states. This Hamiltonian is well known in quantum optics where it describes the non-degenerate optical parametric oscillator. In general it leads to a complicated entanglement between the pump beam ($\hat{c}$) and the signal and idler beams $\hat{a}_0, \hat{a}_1$ which can of course be made subject to detailed investigation [9], but in the limit of a strong pump, the depletion can often be ignored and the pump operators can be replaced by c-numbers. We will assume here that the molecular condensate is sufficiently large that depletion can be neglected, and hence we shall consider the simpler Hamiltonian

$$\hat{H} = \chi \hat{a}_0^\dagger \hat{a}_1^\dagger + \chi^* \hat{a}_0 \hat{a}_1 \tag{2}$$

The coupling strength χ now includes the c-number describing the molecular field. Note that the quadratic Hamiltonian (2) does not conserve the number of atoms, and the final state appears to be a coherent superposition of states with different numbers of atoms. This is, however, only because we omit the meticulous writing of the associated molecular components of the states, which precisely account for the conservation of atom numbers. As we shall only be interested in number conserving observables such as the atomic density distribution, we shall make no errors in applying the symmetry breaking Hamiltonian (2).

Starting in a state with no atoms, the atomic vacuum state, in both modes, the time evolution leads to the production of the two-mode state

$$|\Psi\rangle = \sqrt{1-|s|^2}\sum_n s^n |n,n\rangle, \tag{3}$$

where

$$s = -i\frac{\chi}{|\chi|}\tanh(|\chi|t/\hbar). \tag{4}$$

As discussed in detail in the Introduction, this state does not follow from a mean field analysis and, indeed, the mean values of the field operators $\hat{a}_i$ vanish exactly, whereas the state has a mean atom number of

$$\langle \hat{N}_i \rangle = \langle \hat{a}_i^\dagger \hat{a}_i \rangle = |s|^2/(1-|s|^2) = \sinh^2(|\chi|t/\hbar) \tag{5}$$

in each mode. The atom number distributions are exponential (thermal) and thus the fluctuations in the atom number are large, $\mathrm{Var}(\hat{N}_i) = \langle \hat{N}_i \rangle^2 + \langle \hat{N}_i \rangle$.

A number of papers have discussed observational difference between systems populating several single particle states macroscopically and a coherent superposition of the same states. It has been argued [10, 11, 12, 13] that such differences are small or insignificant for systems with many atoms. The general argument was supplemented by simulations of actual detection records, where the back action due to local measurements on the system turned out precisely to establish a definite relative phase of the two components.

The restriction to only two modes implies that the one-body density matrix is fully characterized by the 2x2 matrix with $\rho_{ij} = \langle a_i^\dagger a_j \rangle$. In particular the spatial density is given by

$$\begin{aligned} n(x,y) = \rho_{00}|\Phi_{00}(x,y)|^2 + \rho_{11}|\Phi_{01}(x,y)|^2 \\ + 2\mathrm{Re}[\rho_{01}\Phi_{00}(x,y)\Phi_{01}^*(x,y)]. \end{aligned} \tag{6}$$

Registration of a particle at position (x_d, y_d), chosen according to this probability distribution, causes the application of the annihilation operator $\Phi_{00}(x_d,y_d)\hat{a}_0 + \Phi_{01}(x_d,y_d)\hat{a}_1$ on the state vector expanded in the two

mode number components as in (3), followed by a renormalization. We are thus able to compute the up-dated 2x2 density matrix and by repeating these steps to simulate the subsequent detection of a number k of atoms. The present problem differs in two way from the simulations reported in Refs. [10, 12]: the two modes populated are not plane waves but they have different spatial dependencies, implying that some atomic detection events can be ascribed solely to one component (e.g., only the $m = 0$ component contributes to the atomic density on the vortex axis), and the inital state does not have a well defined number of atoms, but much larger than Poissonian fluctuations, and hence the rigid mathematical analysis of the emergence of a relative phase in Ref. [11] does not apply. We have carried out simulations, and despite these two differences, the 2x2 density matrix indeed approaches a pure state projector and we obtain detection patterns that are compatible with the density of a single coherent quantum state.

III. MULTI-MODE ANALYSIS

Let us now analyze the problem without making the two-mode simplification. Retaining the approximation of replacing molecule operators with c-numbers, a full many-mode version of Eq.(2) reads

$$H = \int d^3r\ \hat{\Psi}^\dagger(\mathbf{r}) h(\mathbf{r}) \hat{\Psi}(\mathbf{r}) + \left\{ \int d^3r d^3r'\ \chi(\mathbf{r},\mathbf{r}') \hat{\Psi}^\dagger(\mathbf{r}) \hat{\Psi}^\dagger(\mathbf{r}') + \text{h.c.} \right\}. \tag{7}$$

The single particle part consists of kinetic energy and external trapping potential

$$h(\mathbf{r}) = \frac{\mathbf{p}^2}{2m_{\rm atm}} + V_{\rm ex}(\mathbf{r}) - \Delta_{\rm bare}. \tag{8}$$

The energy offset $\Delta_{\rm bare}$ is half of the two-atom detuning: the energy of a trapped molecule in the vortex state is the energy of two free atoms plus $2\Delta_{\rm bare}$.

Typical traps are well approximated by a harmonic potential $V_{\rm ex}(\mathbf{r}) = m_{\rm atm}(\omega_x^2 x^2 + \omega_y^2 y^2 + \omega_z^2 z^2)/2$. We will consider situations with axial symmetry and we therefore let $\omega_x = \omega_y = \omega$. For the purpose of discussing the dissociation of a molecular condensate with a vortex along z, it is useful to make one of two simplifying assumptions about the dynamics along z:

- A quasi cylindrical situation with $\omega_z \ll \omega_\perp$ such that the z component of the momentum, p_z, becomes approximately conserved.
- A quasi two-dimensional situation where $\omega_z \gg \omega_\perp$ so that the dynamics along z is effectively frozen to take place in a single quantum state.

We focus on the second case in the following. The modifications needed to treat the first case are briefly discussed in the Appendix.

The Hamiltonian (7) can describe many processes where pairs of particles are created: one must choose the correct coupling kernel $\chi(\mathbf{r},\mathbf{r}')$ for the problem under consideration. When dealing with dissociation of molecules, $\chi(\mathbf{r},\mathbf{r}')$ depends on the wavefunction of individual molecules (both relative motion and center-of-mass), and on the possible spatial variation of the external fields mediating the dissociation. We will assume the relative motion molecular wavefunction to be very well localized compared to other length scales in the problem. This means that the dependence of $\chi(\mathbf{r},\mathbf{r}')$ on the separation $\mathbf{r}-\mathbf{r}'$ can be approximated by a delta-function. We further assume that the center-of-mass molecular wavefunction and the relevant external fields are rotationally symmetric around the z-axis so that

$$\chi(\mathbf{r},\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}')\chi_{\rm cm}(\rho,z)e^{iM\phi}, \tag{9}$$

in usual cylindrical coordinates where $x = \rho\cos\phi$ and $y = \rho\sin\phi$. In the simplest case the dissociating external fields are spatially uniform over the extend of the molecular condensate. Then M is the charge of the molecular vortex state and the remaining spatial variation of $\chi_{\rm cm}(\rho,z)$ is simply given by the norm of the c-number field that describes the molecular condensate (squareroot of the density) [20]. To specify the overall strength of the coupling a microscopic model of the dissociation must be decided upon. In Ref. [14], a two-photon Raman process between a bound molecular state and "free" (except for external trapping) atoms is considered. The dissociation is via an excited molecular state, which should ideally be only negligibly populated since spontaneous decay from it will be an unwanted loss mechanism. The particular example in Ref. [14] shows that a peak value of our χ comparable to the trap level spacing is fully realistic. This is the regime we focus on below.

We emphasize that this physical modelling does not favor any particular pair of atomic modes, and the Hamiltonian (7) cannot be brought on the form (2). Atoms are coherently prepared from the entire region populated by the molecules, but pairs are initially much more tightly located both radially and azimuthally than the single mode functions suggested in the previous section. In particular, the atoms are not restricted to low values of the angular momentum quantum number. The temporal evolution of the system due to the kinetic energy and external potential, and the role of energy conservation, however, favors the population of only few modes as we shall see below.

A. Frozen z dynamics

In the case of tight z confinement for the atoms, we get a simple description. Let the only accessible atomic z mode be ϕ_z, $\int dz|\phi_z|^2 = 1$. We can then expand the atomic field operators on the discrete set of mode opera-

tors

$$\hat{\Psi}(\mathbf{r}) = \sum_{nm} \hat{a}_{nm} \times \Phi_{nm}(\rho) \times \sqrt{\frac{1}{2\pi}} e^{im\phi} \times \phi_z(z) \quad (10)$$

where the radial modefunction Φ_{nm} are

$$\begin{aligned}\Phi_{nm}(\rho) = &\sqrt{\frac{n!}{2\,(n+|m|)!a_{\rm osc}^2}} \\ &\times \exp\left(-\frac{1}{2}\frac{\rho^2}{a_{\rm osc}^2}\right)\left(\frac{\rho}{a_{\rm osc}}\right)^{|m|} L_n^{|m|}\left(\frac{\rho^2}{a_{\rm osc}^2}\right),\end{aligned} \quad (11)$$

with L_n^m the m'te associated Laguerre polynomum of order n and $a_{\rm osc} = \sqrt{\hbar/m_{\rm atm}\omega}$ is the oscillator length. The commutation relations of the $\hat{a}_{nm}$ are

$$\begin{aligned}\left[\hat{a}_{nm}, \hat{a}_{n'm'}\right] &= 0 \\ \left[\hat{a}_{nm}, \hat{a}^\dagger_{n'm'}\right] &= \delta_{nn'}\delta_{mm'}.\end{aligned} \quad (12)$$

The Hamiltonian becomes

$$\begin{aligned}\hat{H} = &\sum_{nm} E_{nm}\hat{a}^\dagger_{nm}\hat{a}_{nm} \\ &+ \sum_m \sum_{nn'} \left\{K_{nn'm}\hat{a}^\dagger_{nm}\hat{a}^\dagger_{n'(M-m)} + \text{h.c.}\right\},\end{aligned} \quad (13)$$

where $K_{nn'm}$ is defined via

$$\begin{aligned}&K_{nn'm} = \\ &\int dz d\rho\rho\ \chi_{\rm cm}(\rho, z)\ |\phi(z)|^2\ \Phi^*_{nm}(\rho)\Phi_{n'M-m}(\rho).\end{aligned} \quad (14)$$

and E_{nm} is given by

$$E_{nm} = (2n + |m| + 1)\,\hbar\omega - \Delta. \quad (15)$$

Here Δ is the effective detuning, adjusted for the energy associated with ϕ_z:

$$\Delta = \Delta_{\rm bare} - \int dz\ \phi_z^*\left(\frac{p_z^2}{2m} + \frac{1}{2}m\omega_z^2 z^2\right)\phi_z. \quad (16)$$

IV. BOGOLIUBOV DIAGONALIZATION

The Hamiltonian (13) is a quadratic form of creation and annihilation operators and as such it can in general be decoupled to a collection of independent harmonic oscillators by a *Bogoliubov transformation*. Please note that this transformation is usually applied in connection with the *Bogoliubov approximation* in theoretical studies of Bose-Einstein condensed systems. The Bogoliubov approximation provides a quadratic Hamiltonian by appeal to the macroscopic population of the condensate mode. Here the quadratic form of the Hamiltonian is rather a consequence of (i) that two atoms are created in each fundamental dissociation process and (ii) that we ignore the dynamics (in particular the depletion) of the molecules and describe them by a stationary c-number field. One could in principle extend our approach to also treat the interaction among the created atoms in a Bogoliubov approximation, but since we focus on situations with initially *no* atoms present this would neither be necessary nor indeed well justified. For an example of *time-dependent* quadratic Hamiltonians, see the work by Ziń *et al.* on colliding condensates [15, 16].

A. Two-mode Bogoliubov

Let us first look at the simple case of two modes like in Sec. II. In addition to the pair creation and annihilation terms of Eq. (2) we include mode energies $\hbar\omega_0$ and $\hbar\omega_1$:

$$\begin{aligned}\hat{H} = &\hbar\omega_0\left(\hat{a}_0^\dagger\hat{a}_0 + \frac{1}{2}\right) + \hbar\omega_1\left(\hat{a}_1^\dagger\hat{a}_1 + \frac{1}{2}\right) \\ &+ \chi\hat{a}_0^\dagger\hat{a}_1^\dagger + \chi^*\hat{a}_0\hat{a}_1.\end{aligned} \quad (17)$$

A useful and compact notation is

$$\hat{H} = \frac{1}{2}\hat{A}^\dagger \mathrm{h}\hat{A} \quad (18)$$

with

$$\hat{A} = \begin{bmatrix}\hat{a}_0 \\ \hat{a}_1 \\ \hat{a}_0^\dagger \\ \hat{a}_1^\dagger\end{bmatrix}, \quad A^\dagger = \begin{bmatrix}\hat{a}_0^\dagger & \hat{a}_1^\dagger & \hat{a}_0 & \hat{a}_1\end{bmatrix}, \quad (19)$$

and

$$\mathrm{h} = \begin{bmatrix}\hbar\omega_0 & 0 & 0 & \chi \\ 0 & \hbar\omega_1 & \chi & 0 \\ 0 & \chi^* & \hbar\omega_0 & 0 \\ \chi^* & 0 & 0 & \hbar\omega_1\end{bmatrix}. \quad (20)$$

We now seek to simplify $\hat{H}$ by a Bogoliubov transformation, i.e. we define new creation and annihilation operators implicitly by

$$\begin{bmatrix}\hat{a}_0 \\ a_1 \\ \hat{a}_0^\dagger \\ a_1^\dagger\end{bmatrix} = \begin{bmatrix}U & V^* \\ V & U^*\end{bmatrix}\begin{bmatrix}\hat{b}_0 \\ \hat{b}_1 \\ \hat{b}_0^\dagger \\ \hat{b}_1^\dagger\end{bmatrix} \quad (21)$$

where the 2×2 matrices U and V should fulfill

$$\begin{aligned}U^\dagger U - V^\dagger V &= \mathbb{I}_2 \\ U^{\rm T}V - V^{\rm T}U &= 0\end{aligned} \quad (22)$$

in order for $\hat{b}_0$ and $\hat{b}_1$ to have the commutation relations for independent bosonic creation and annihilation operators [see Eq. (12)]. Note that in general the Bogoliubov transform is *not* simply a choice of different spatial modes.

1. Detuning dominated case $2|\chi| < |\hbar\omega_0 + \hbar\omega_1|$

Depending on the strength of the coupling $|\chi|$ relative to the two-atom detuning $|\hbar\omega_0 + \hbar\omega_1|$ the Hamiltonian can be written in one of two standard forms. If $2|\chi| < |\hbar\omega_0+\hbar\omega_1|$, $\hat{H}$ can be written as a sum of two independent oscillators

$$\hat{H} = \lambda_0 \left(\hat{b}_0^\dagger \hat{b}_0 + \frac{1}{2}\right) + \lambda_1 \left(\hat{b}_1^\dagger \hat{b}_1 + \frac{1}{2}\right), \tag{23}$$

with

$$\begin{aligned}\lambda_{0,1} =& \pm \frac{1}{2}(\hbar\omega_0 - \hbar\omega_1) \\ &+ \frac{1}{2}(\hbar\omega_0 + \hbar\omega_1)\sqrt{1 - \frac{4|\chi|^2}{|\hbar\omega_0 + \hbar\omega_1|^2}}\end{aligned} \tag{24}$$

The Bogoliubov transformation is given by:

$$U = \begin{bmatrix} \cosh r & 0 \\ 0 & \cosh r \end{bmatrix} \tag{25}$$

and

$$V = \begin{bmatrix} 0 & \mp\frac{\chi^*}{|\chi|}\sinh r \\ \mp\frac{\chi^*}{|\chi|}\sinh r & 0 \end{bmatrix}, \tag{26}$$

where the upper (lower) sign should be chosen for $\hbar\omega_0 + \hbar\omega_1$ positive (negative). The squeezing parameter r is defined through

$$\tanh r = \frac{2|\chi|}{|\hbar\omega_0 + \hbar\omega_1| + \sqrt{|\hbar\omega_0 + \hbar\omega_1|^2 - 4|\chi|^2}} \tag{27}$$

We see that as $2|\chi|$ approaches $|\hbar\omega_0 + \hbar\omega_1|$, the coefficients in the transformation diverge, i.e. the decoupled modes become infinitely squeezed.

Because of the diagonalized form of the Hamiltonian (23), the time evolution will be independent for the two Bogoliubov modes: Each will simply behave as a harmonic oscillator. This means that the dynamics of all quantities will be oscillatory with two fundamental frequencies given by $\lambda_{0,1}/\hbar$.

Note, that even if both mode energies $\hbar\omega_{0,1}$ are positive so that a pair of atoms actually has a higher energy than a molecule, one of the eigenvalues λ_i can become negative. This happens if $|\chi|^2 > \hbar\omega_0\hbar\omega_1$ and in that case the system will be thermodynamically unstable as it is energetically favourable to create the corresponding kind of quasi-particles.

2. Coupling dominated case $2|\chi| > |\hbar\omega_0 + \hbar\omega_1|$

If $2|\chi| > |\hbar\omega_0 + \hbar\omega_1|$, the simplest form attainable by Bogoliubov transformations involves pair-creation (with real positive coefficient) into two symmetrically detuned modes:

$$\hat{H} = \mathrm{Re}[\lambda]\left(\hat{b}_1^\dagger \hat{b}_1 - \hat{b}_0^\dagger \hat{b}_0\right) + \mathrm{Im}[\lambda]\left(\hat{b}_0^\dagger \hat{b}_1^\dagger + \hat{b}_0 \hat{b}_1\right) \tag{28}$$

with

$$\begin{aligned}\lambda =& \frac{1}{2}(\hbar\omega_1 - \hbar\omega_0) \\ &+ \frac{i}{2}(\hbar\omega_0 + \hbar\omega_1)\sqrt{\frac{4|\chi|^2}{|\hbar\omega_0 + \hbar\omega_1|^2} - 1}.\end{aligned} \tag{29}$$

The transformation is still of the form given in Eqs. (25) and (26), but now the squeezing parameter r should be found from:

$$\tanh r = \frac{|\hbar\omega_0 + \hbar\omega_1|}{2|\chi| + \sqrt{4|\chi|^2 - |\hbar\omega_0 + \hbar\omega_1|^2}} \tag{30}$$

We see that as before, r diverges when $2|\chi|$ approaches $|\hbar\omega_0 + \hbar\omega_1|$. At exact two-*atom* resonance, i.e. $\hbar\omega_0 + \hbar\omega_1 = 0$, the Bogoliubov operators are simply the original $\hat{a}_0$ and $\hat{a}_1$.

The transformed Hamiltonian (28) consist of two commuting terms and the first one even conserves the number of quasi-particles $\hat{N}_b = \hat{b}_0^\dagger \hat{b}_0 + \hat{b}_1^\dagger \hat{b}_1$. The second term leads to unbound creation of quasi-particle pairs, in fact $\langle \hat{N}_b \rangle$ will grow as $\sinh^2(\mathrm{Im}[\lambda]t)$. This production of quasi-particles translates to a similarly unbounded growth in the number of atoms (until depletion of the molecular condensate renders our simple Hamiltonian invalid), and because of the Bogoliubov tranformation, the atom numbers will show oscillations around their general growth.

B. Multi-mode Bogoliubov

The generalization to many modes is relatively straightforward. By Bogoliubov transformations of the original set of creation and annihilation operators, $\hat{H}$ can be written as a sum of a number of independent oscillators and a number of pairs of symmetrically detuned modes into which quasi-particles are created:

$$\begin{aligned}\hat{H} =& \sum_j \lambda_j \left(\hat{b}j^\dagger \hat{b}_j + \frac{1}{2}\right) \\ &+ \sum_p \mathrm{Re}[\lambda_p]\left(\hat{b}_{p2}^\dagger \hat{b}_{p2} - \hat{b}_{p1}^\dagger \hat{b}_{p1}\right) \\ &+ \mathrm{Im}[\lambda_p]\left(\hat{b}_{p1}^\dagger \hat{b}_{p2}^\dagger + \hat{b}_{p1}\hat{b}_{p2}\right).\end{aligned} \tag{31}$$

The dynamics in the decoupled modes is oscillatory while ever more quasi-particles will be created in the paired modes. Note that it is perfectly possible that all modes can be decoupled so that the unitary dynamics is purely oscillatory.

C. Consequences of the azimuthal symmetry

We consider a situation with axial symmetry: The single particle part of the Hamiltonian, $h(\mathbf{r})$ of Eq. (8), is invariant under rotations around the z-axis. This leads to the usual block-diagonal form of the single particle Hamiltonian with respect to the quantum number m, the eigenvalue of the z-component of the angular momentum operator. At the same time, the molecular field [χ of Eq. (9)] changes simply by a phase factor $\exp(iM\phi)$ when the system is rotated by an angle ϕ. This leads to an exact selection rule on the pairs of modes that are populated in the dissociation process, namely $m + m' = M$. For odd M this imposes an "off-diagonal" form on the pair creation, as the atoms created by a dissociation process must necessarily end up with different integer quantum numbers, m and $M - m$, whereas for even M the two atoms can both end up in an $m = M/2$ mode.

In summary, each m subspace is coupled only to itself by the single particle part of the Hamiltonian and the dissociation can either couple it exclusively to itself or exclusively to one other m subspace. In this way the full problem splits into a series of smaller problems consisting of either one or two m-subspaces.

Now consider the problem of two coupled subspaces. As pairs are always created with an atom in each subspace, there is an obvious conservation of the *difference* in the total number of atoms in the two subspaces. This can also be phrased as the invariance of the evolution under shifts in the *relative* energy of the two subspaces as long as their total energy is maintained. Among other consequences, this implies that there are no first-order coherences between the two subspaces and that the one-body density operator is correspondingly block-diagonal. It is, in fact, possible to show an even stronger, dynamical symmetry in the evolution: If the two subspaces are initially unpopulated, they will at all times remain unitarily equivalent. In particular, their one-body density matrices will have identical spectra of eigenvalues [17].

V. RESULTS

As discussed above, once the Hamiltonian has been brought to a standard form by a Bogoliubov transformation, a number of properties of the system are immediately clear. In particular, the oscillatory or unbounded behaviour depends on whether all modes can be decoupled. In Fig. 1 we show the real and imaginary parts of the λ_j's as a function of the detuning Δ. When Δ is increased, all atomic modes move linearly down in energy according to the single atom Hamiltonian (7). As a pair of modes with $m + m' = M = 1$ gets close to the two-atom resonance, i.e. to having equal and opposite energies, the coupling has the effect of bending the quasi-particle energy levels further downwards. The modes become increasingly squeezed but remain uncoupled – they are in the detuning dominated regime of Sec. IV A 1. At

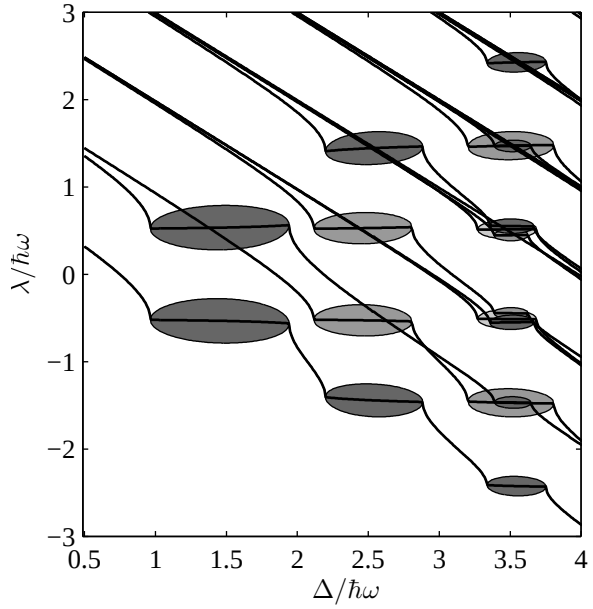

FIG. 1: Bogoliubov eigenvalues as functions of the detuning Δ for the Hamiltonian (13) with a coupling field (9) with $M = 1$. For concreteness, we assume a strength and a radial dependence of the coupling such that $\int dz \chi_{\rm cm}|\phi_z|^2 = 2\hbar\omega\rho\exp(-\rho^2/a_{\rm osc}^2)/a_{\rm osc}$. The lines show $\mathrm{Re}(\lambda)$, which are the mode energies while, in the coupling dominated regime, the shaded areas around them have a height of $\mathrm{Im}(\lambda)$ to indicate the coupling between pairs of modes. The darkness of the shading indicates the m-values in the pairs of coupled modes: the darkest shading is for $m = 0, m' = 1$ pairs, $m = -1, m' = 2$ pairs are a tone brighter, etc. Note that members of coupled pairs lie symmetrically around $\lambda = 0$.

the point of perfect two-quasi-particle resonance, it is no longer possible to decouple the two modes and the system moves into a regime with quasi-particle pair production, the coupling dominated case of Sec. IV A 2. When a pair of quasi-particle modes become unstable, it is signalled by the corresponding λ_j's attaining a finite imaginary part. In Fig. 1 this is plotted as a shaded region around the corresponding energy curves. The height of the shaded region signifies the magnitude of $\mathrm{Im}[\lambda]$. As Δ is further increased, the pair production strength also increases. It has its maximum approximately at the point of two-*atom* resonance, i.e., when the two *bare* atomic modes becomes resonant with the molecule energy.

As an example, let us follow the lowermost curve through the figure. At the left, at low values of Δ, the curve corresponds to the atomic harmonic oscillator ground state in the trap with $n = 0$ and $m = 0$. When Δ gets close to 1, pair production together with the $(n = 0, m = 1)$ state (second lowest curve) becomes resonant. Note that the $(n = 0, m = 1)$ state is degenerate with the $(n = 0, m = -1)$ for very low Δ. At approximately $\Delta = 1.5\hbar\omega$ [i.e. $E_{00} + E_{01} = 0$, cf. Eq. (15)] the pair production has its maximum. A small shift from the simple estimate is due to the multi-mode character of the

problem. The next resonance for the $(n = 0, m = 0)$ level is centered around at $\Delta = 2.5\hbar\omega$ [i.e. $E_{00} + E_{11} = 0$]. Note that around $\Delta = 2.5\hbar\omega$, the production of pairs with $(n = 0, m = -1)$ and $(n = 1, m = 2)$ is also resonant. The third resonance, where $E_{00} + E_{21} = 0$, is at $\Delta = 3.5\hbar\omega$. Here several other resonances are also present.

In Fig. 2 we plot the total number of atoms and in Fig. 3 the fractional distribution on single-particle modes as a function of time for three different values of Δ. For

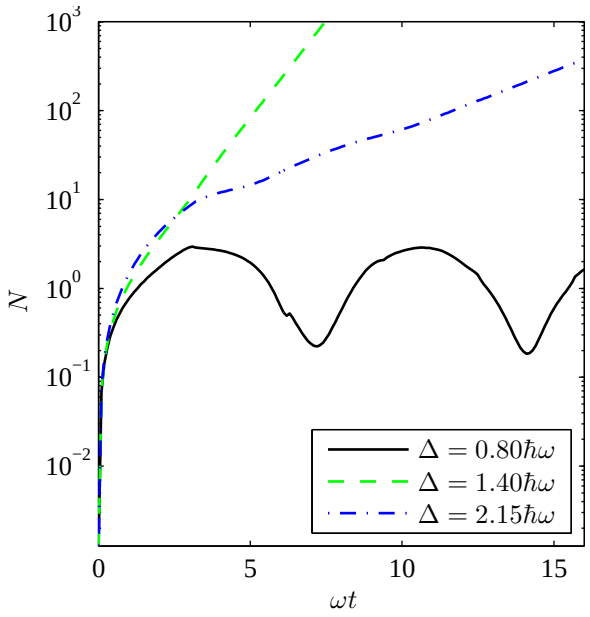

FIG. 2: (color online) Total number of atoms as a function of time for three values of Δ. The coupling is as in Fig. 1.

$\Delta = 0.80\hbar\omega$ the dynamics is detuning dominated and oscillatory. All modes can be decoupled and the resulting quasi-particle modes are only slightly squeezed resulting in a very modest production of atoms. This almost vanishing population mainly occupies two modes: one in the $m = 0$ manifold and one in the $m' = 1$ manifold. These modes constitute the pair that is closest to resonance and their oscillation period is of the same order of magnitude as the trap period. The fractional occupation of these modes is close to 50% except for times close to the minima in their oscillation (e.g. $\omega t \sim 7$ here). At $\Delta = 1.40\hbar\omega$, the system can no longer be decoupled: A single $m = 0$, $m' = 1$ pair of modes has just become unstable and real pair production takes place. Therefore this pair of modes quickly becomes dominant. Finally, for $\Delta = 2.15\hbar\omega$ the evolution is more complex. From Fig. 1 one can tell that there will be real pair production into a pair of modes with $m = -1$, $m' = 2$. However, a pair of modes with $m = 0, m' = 1$ also becomes unstable if Δ is increased just a little bit. It turns out that the $m = 0, m' = 1$ pair dominates at early times, but later most atoms are produced in the $m = -1, m' = 2$ pair of states.

The above results all concern one-body properties. An

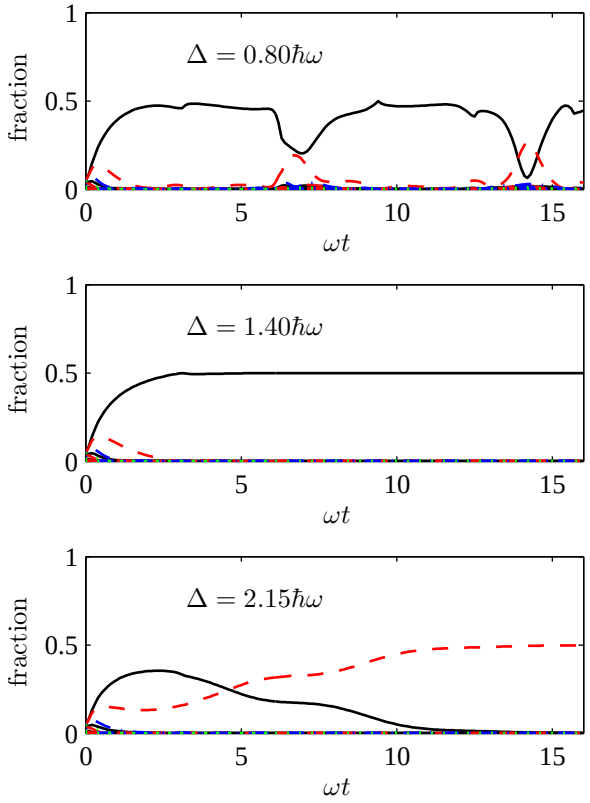

FIG. 3: (color online) Condensate fraction, i.e. relative weigth of the largest eigenvalue of the one-body density operator compared to the sum of all eigenvalues (total number of particles. Results are plotted for three different values of Δ. As discussed in Sec. IV C, the conservation of angular momentum dictates a block-diagonal form of the one-body density operator and that pairs of blocks for which $m + m' = M$ will have identical spectra. Fully drawn lines correspond to $m = 1$ and $m' = 0$, dashed lines to $m = 2$ and $m' = -1$, and dotted lines to $m = 3$ and $m' = -2$.

illustrative way to quantify some of the two-body correlations in the system is to calculate the *conditional* density distribution, i.e., the density distribution resulting after the detection of a single atom at some position (x_d, y_d). In Fig. 4 we plot the result of such a calculation for $\Delta = 2.15\hbar\omega$ and $\omega t = 2, 4, 6$. The upper row shows the normalized density distribution, that is, the probability distribution for the detection of the first atom. In the lower row, an atom has been detected at $(x_d, y_d) = (1, 0)a_{\rm osc}$ and the resulting conditional probability distribution for the next detection is shown. For all three times, this conditional distribution is dramatically different from the original one. Close to (x_d, y_d), there is generally a significantly higher probabilty density, reminiscent of the familiar (thermal state) bunching of bosons. However, when higher angular momentum states are populated, additional peaks appear as the detection induces coherences between more m values. From a calculation of the full one-body density operator we can also find the new "condensate fraction" and for all three

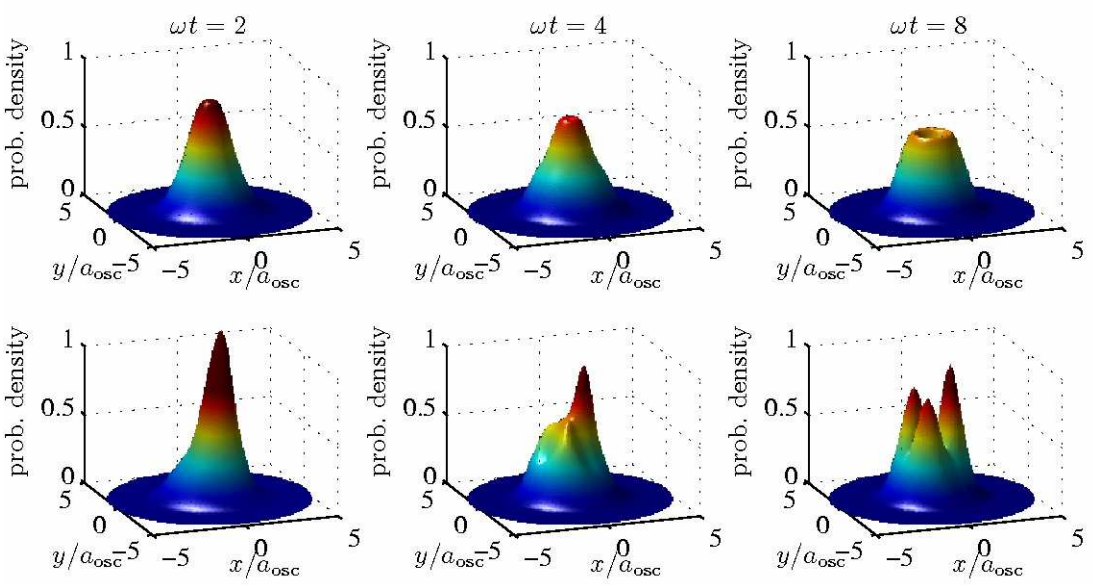

FIG. 4: (color online) Detection probability distribution (normalized density distribution) before (upper row) and after (lower row) detection of a single atom in $(x_d, y_d) = (1, 0)a_{\rm osc}$. The detuning Δ was chosen to be $2.15\hbar\omega$. The three columns show results where the system was allowed to evolve for a time $\omega t = 2, 4$, and 8, respectively. Other parameters were like in the previous figures. Before the detection of the first atom, the density distribution is rotationally symmetric and it is equally likely to make the first detection at any azimuthal angle, ϕ. According to Fig. 3, the one-body density matrix at $\omega t = 2$ is an almost equal mixture of $m = 0$ and $m' = 1$ atoms. In each of these m-manifolds, a single radial mode dominates and accounts for 35% of the total number of atoms. In the lower panel, we assume that an atom has been detected with $\phi = 0$, more precisely at $(x_d, y_d) = (1, 0)a_{\rm osc}$. The conditional probability distribution for the next detection has become asymmetric indicating a significantly increased probability to detect the next atom close to the first one. Diagonalization shows that the one-body density matrix now has a "condensate fraction" of almost 60%. At $\omega t = 4$ a significant number of $m = -1, m' = 2$ pairs have been created. The density before detection has a pronounced dip at the center due to centrifugal effects. The conditional density after the detection is rather complex: it is in fact not even symmetric around the y-axis. Because of the larger number of modes involved, the condensate fraction is only 43%. Finally, at $\omega t = 8$ the $m = -1, m' = 2$ pairs dominate. After the detection, three almost equally strong peaks are seen, signaling the appearence of coherences between two modes with angular momentum differing by 3 units. The condensate fraction after the detection is now again almost 60%.

cases this number is significantly increased compared to the situation before the detection (cf. caption of Fig. 4). This is similar to what has been proposed in the case of number state condensates [18].

VI. CONCLUSION

We have presented a qualitative discussion and a quantitative analysis of the break-up of a molecular condensate into an atomic system, in the case where the molecules occupy a vortex state with unit angular momentum. The rotational symmetry of the problem causes the fragmentation of the atomic system into (at least) two spatial components, and our multi-mode analysis quantified the dynamics of these components. Particle detection at random locations will generally break the rotational symmetry of the state, and both our simple two-mode model and our more general analysis show that such detections will indeed cause the system to approach a single macroscopically populated quantum state with a preferred angular dependence.

Several interesting possibilities exist for further studies. A more technical issue concerns our assumption of non-interacting atoms. As shown in [7] interactions have non-trivial consequences for the quantum correlations in the system. Depletion of the molecular condensate [14] affects the quantum statistics of the atomic state, and moreover, the explicit inclusion of the molecular field will also enable processes where atoms in different fragments recombine and form molecules with new angular momenta, respecting always the overall rotational symmetry of the system, but introducing multi-atom correlations in the system. Within our simple model with non-interacting atoms and undepleted molecular systems, it is a natural question to ask, what atomic states will result from the dissociation of a molecular vortex *lattice*. Will two fragments be sufficient to describe such states? We assumed a cylindrical symmetry and referred extensively to angular momentum conservation in the arguments of this paper, but in an *interacting* system vortices also exist under non-symmetric confinement, and our symmetry argument should be adequately modified into an argument referring to the phase topology. Presumably one

would still see the formation of a fragmented system with components with and without vorticity, but the need for interactions among the atoms for the stability of such a state is an interesting issue.

Acknowledgments

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APPENDIX: CYLINDRICAL SYMMETRY

The case of strict cylindrical symmetry can be treated in a very similar fashion as the one of frozen z dynamics of Sec. III A. Assume that potential is axially symmetric and flat along z: $\omega_z = 0$. The coupling is also assumed to be z independent, $\chi_{\rm cm}(\rho, z) = K(\rho)$. We then use a quantization box of length $2L$ along z for the atoms. The field operator is expanded as follows

$$\hat{\Psi}(\mathbf{r}) = \sum_{n=0}^{\infty}\sum_{m=-\infty}^{\infty}\sum_{k=-\infty}^{\infty} \hat{a}_{nmk} \times \Phi_{nm}(\rho) \times \sqrt{\frac{1}{2\pi}} e^{im\phi} \times \sqrt{\frac{1}{2L}} e^{i\pi kz/L} \quad \text{(A.1)}$$

where the radial modefunctions where given in Eq. (11). The mode creation and annihilation operators satisfy usual commutation relations

$$\begin{aligned} \left[\hat{a}_{nmk}, \hat{a}_{n'm'k'}\right] &= 0 \\ \left[\hat{a}_{nmk}, \hat{a}^{\dagger}_{n'm'k'}\right] &= \delta_{nn'}\delta_{mm'}\delta_{kk'}. \end{aligned} \quad \text{(A.2)}$$

Under the above assumptions, the Hamiltonian (7) then simplifies to the form:

$$\hat{H} = \sum_{nmk} E_{nmk}\hat{a}^{\dagger}_{nmk}\hat{a}_{nmk} + \sum_{mk}\sum_{nn'}\left\{K_{nn'm}\hat{a}^{\dagger}_{nmk}\hat{a}^{\dagger}_{n'(M-m)(-k)} + \text{h.c.}\right\}, \quad \text{(A.3)}$$

where

$$E_{nmk} = 2n + |m| + 1 + \frac{1}{2}\left(\frac{\pi}{L}\right)^2 k^2 - \Delta \quad \text{(A.4)}$$

and

$$K_{nn'm} = \int dz d\rho\rho\; K(\rho)\; \frac{1}{2L}\; \Phi^{*}_{nm}(\rho)\Phi_{n'M-m}(\rho). \quad \text{(A.5)}$$

The main difference to the case of a single active z mode treated above is that excess molecular energy can now be transformed to z kinetic energy of the atoms. Thus the picture of isolated resonances breaks down, even at low coupling strengths. Many atomic modes will participate in the dynamics and it will take more detections to build up a sizable condensate fraction.

[1] M. R. Andrews, C. G. Townsend, H.-J. Miesner, D. S. Durfee, D. M. Kurn, and W. Ketterle, Science **275**, 637 (1997).
[2] K. W. Madison, F. Chevy, W. Wohlleben, and J. Dalibard, Phys. Rev. Lett. **84**, 806 (2000).
[3] M. W. Zwierlein, J. R. Abo-Shaeer, A. Schirotzek, C. H. Schunck, and W. Ketterle, Nature **435**, 1047 (2005).
[4] N. Gemelke, E. Sarajlic, Y. Bidel, S. Hong, and S. Chu, Phys. Rev. Lett. **95**, 170404 (pages 4) (2005).
[5] G. K. Campbell, J. Mun, M. Boyd, E. W. Streed, W. Ketterle, and D. E. Pritchard, Phys. Rev. Lett. **96**, 020406 (pages 4) (2006).
[6] K. M. Hilligsøe and K. Mølmer, Phys. Rev. A **71**, 041602(R) (pages 4) (2005).
[7] U. V. Poulsen and K. Mølmer, Phys. Rev. A **63**, 023604 (2001).
[8] K. V. Kheruntsyan, M. K. Olsen, and P. D. Drummond, Phys. Rev. Lett. **95**, 150405 (2005).
[9] K. Dechoum, P. D. Drummond, S. Chaturvedi, and M. D. Reid, Phys. Rev. A **70**, 053807 (pages 24) (2004).
[10] J. Javanainen and S. M. Yoo, Phys. Rev. Lett. **76**, 161 (1996).
[11] Y. Castin and J. Dalibard, Phys. Rev. A **55**, 4330 (1997).
[12] K. Mølmer, Phys. Rev. A **55**, 3195 (1997).
[13] J. I. Cirac, C. W. Gardiner, M. Naraschewski, and P. Zoller, Phys. Rev. A **54**, R3714 (1996).
[14] D. J. Heinzen, R. Wynar, P. D. Drummond, and K. V. Kheruntsyan, Phys. Rev. Lett. **84**, 5029 (2000).
[15] P. Zin, J. Chwedenczuk, A. Veitia, K. Rzazewski, and M. Trippenbach, Phys. Rev. Lett. **94**, 200401 (pages 4) (2005).
[16] P. Zin, J. Chwedenczuk, and M. Trippenbach, Phys. Rev. A **73**, 033602 (pages 10) (2006).
[17] U. V. Poulsen, in preparation.
[18] K. Mølmer, Phys. Rev. A **65**, 021607(R) (2002).
[19] M. F. Andersen, C. Ryu, P. Clade, V. Natarajan, A. Vaziri, K. Helmerson, and W. D. Phillips, Phys. Rev. Lett. **97**, 170406 (pages 4) (2006).
[20] In principle, the dissociating fields could also contribute to M. Transfer of (orbital) angular momentum in Bragg scattering of BECs has been demonstrated in Ref. [19].